

Symmetry and dimension of the magnon dispersion of inorganic spin-Peierls systems

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The data on the dispersion of the magnetic excitations of CuGeO_3 in the spin-Peierls D-phase are analyzed. On the basis of the lattice structure it is shown that even along the chains the $d = 2$ character cannot be neglected. The symmetry of the dispersion differs from the one assumed so far. The magnetic resonance data is reinterpreted. The possibility of interchain rather than intrachain frustration is discussed.

75.40.Gb, 75.10.Jm, 75.50.Ee

The substance CuGeO_3 is the first inorganic example of a spin-Peierls system [1]. A second compound which is intensively investigated is $\alpha'\text{-NaV}_2\text{O}_5$ [2,3]. An inorganic sample is particularly interesting since it allows to employ all the experimental tools (for a review, see [4]). Thus the investigation of CuGeO_3 has become a very active field both in theory and in experiment.

A spin-Peierls system is an antiferromagnetic spin system coupled to phonons such that below a certain temperature T_{SP} the translational symmetry is spontaneously broken. The system becomes dimerized due to a static lattice distortion which renders the spin coupling alternating along a certain axis. There are stronger bonds (J) and weaker bonds (λJ , $\lambda < 1$). The phenomenon is essentially driven by $d = 1$ physics due to the extreme sensitivity of a $d = 1$ system towards a $2k_F$ perturbation. This is manifest in the superquadratic growth of the energy gain $\Delta E \propto -\delta^{4/3}$ with the dimerization $\delta := (1 - \lambda)/(1 + \lambda)$ [5].

It is the aim of the present work to show that CuGeO_3 is only approximately a $d = 1$ compound. On the basis of the dispersion perpendicular to the chains this has been noted by several authors (e.g. [6–9,4]). What has passed unnoticed so far is that the spin-spin couplings perpendicular to the chains, which are formed along the c -axis, influences the dispersion *along* the chains in a non-trivial, non-constant way.

So far, the deviations of the behavior of CuGeO_3 from the physics of simple nearest-neighbor (NN) coupled Heisenberg chains plus phonons were attributed to an intrachain frustration, i.e. an additional next-nearest-neighbor (NNN), antiferromagnetic coupling J_2 [10–13,9]. But no consensus has been reached which value the relative frustration $\alpha = J_2/J$ [14] applies for CuGeO_3 . The values range from $\alpha = 0.36$ [11] to $\alpha = 0.17$ [9] and the values for J from 10.4 meV [6] to 21.5 meV [15]. This poses a problem since the NNN coupling represents a marginal perturbation which engenders above the critical value $\alpha_c = 0.241$ [16] a gap in the

spin system itself without alternating coupling constants. The high temperature data for the magnetic susceptibility $\chi(T)$ [1,17] suggests values above α_c [11,26] whereas there is no indication for a spin gap without coupling alternation [12]. An analysis of the dispersion $\omega(q_c)$ at small q_c suggests $\alpha = 0.17 < \alpha_c$ [9].

The dispersion in b -direction is roughly 0.3 of the one in c -direction; the dispersion in a -direction is roughly 1/50. First, we focus on the situation in the bc -plane. Up to now, the complete dispersions $\omega(\vec{q})$ is interpreted, at least qualitatively, as being a sum of two independent contributions $\omega(\vec{q}) \approx \omega_b(q_b) + \omega_c(q_c)$ leading to the schemes in [18] (inset fig. 2) and in [4] (fig. 9).

On the other hand, elastic neutron scattering [19,20] provides evidence that the dimerization pattern is shifted by one spacing from one chain to the other, i.e. adjacent copper ions move in opposite directions [19]. A segment of the resulting dimer distribution is shown in fig. 1.

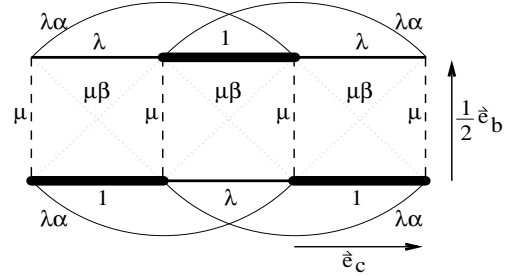


FIG. 1. Segment of the dimerized lattice in the bc -plane. The largest coupling is set to unity and the weaker couplings are parametrized as indicated.

The bold lines represent the dimers (J is set to unity); the other lines (thin, dashed, dotted) represent the residual, weaker couplings ($\lambda, \mu < 1$). The line λ is the weak bond in the dimerized chain, μ is the interchain coupling, $\mu\beta$ is a possible NNN hopping, and $\lambda\alpha$ is the intrachain frustration discussed before. The microscopic superexchange paths for μ and $\mu\beta$ are Cu-O2-O2-Cu in b -direction, see [20] fig. 1. Since from one of the O2 two Cu with different c -coordinate can be reached, a direct super-exchange (μ) or a super-exchange with a shift along the c -axis ($\mu\beta$) is possible [20]. One expects that $\beta \approx 0.5$ because there are two paths for the direct super-exchange and one for each shifted super-exchange.

The analysis starts from the limit of strong dimerization. The experimental systems are closer to $\delta = 0$ than to $\delta = 1$. But the $\delta = 1$ case is a good starting point due to its simplicity which allows a systematic perturbative approach, in principle possible down to arbitrarily

small $\delta > 0$. In practice a third order calculation yields reasonable results down to $\delta \approx 0.1$ (see fig. 1 in [21]).

At $\delta = 1$, the ground state is a product of local singlets on the dimers and a magnon is a degenerate local single triplet. These triplets acquire a dispersion due to the residual couplings which lift the degeneracy. To first order one finds a hopping from dimer to dimer along the chains of $t_{10} = -\lambda(1 - 2\alpha)/4$ [21] and of $t_{11} = -\mu(1 - 2\beta)/4$ to the nearest dimers in b-direction. To obtain the corresponding dispersion one may view the dimers as sites of the effective lattice in fig. 2.

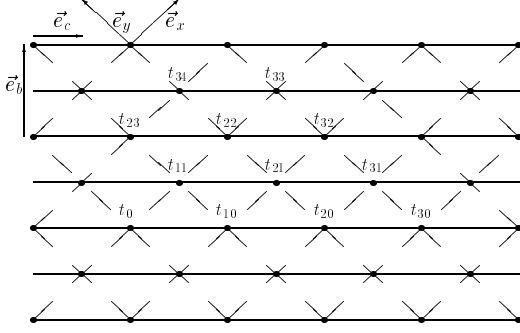


FIG. 2. Effective lattice if the dimers are taken as elementary sites. The hopping elements refer to hops starting at the site t_0 .

Dashed lines correspond to the processes $\propto \mu$ and solid lines to processes $\propto \lambda$. Starting from t_0 one reaches the sites t_{10} , t_{11} and their symmetry equivalents (reflection at the c-axis and the b-axis) with the amplitudes given above. A natural unit cell is spanned by \vec{e}_x and \vec{e}_y as indicated. The dashed lines form a square lattice to which the solid lines add a component along one of the diagonals. The dispersion reads $\omega(\vec{q}) = 2t_{11}(\cos(q_x) + \cos(q_y)) + 2t_{10}\cos(q_x - q_y)$ where $q_{x/y}$ are the components along the dual basis vectors belonging to \vec{e}_x and to \vec{e}_y , respectively. Changing to the basis dual to $\{\vec{e}_b, \vec{e}_c\}$ by $q_{x/y} = q_b/2 \pm q_c$ one obtains

$$\frac{\omega(\vec{q})}{J} = 1 + 2t_{10} \cos(2q_c) + 4t_{11} \cos(q_b/2) \cos(q_c). \quad (1)$$

Eq. (1) illustrates already the main points of this paper. A dispersion of type (1) has minima at $\vec{q} = (0, 0, 0)$ and at $(0, 1, 1/2)$. Saddle points are at $(0, 1, 0)$ and at $(0, 0, 1/2)$. These features stay valid in higher perturbative orders as well. They change the picture used so far (cf. fig. 9 in [4]) where the symmetry relations $(0, 0, 0) \leftrightarrow (0, 0, 1/2)$ and $(0, 1, 0) \leftrightarrow (0, 1, 1/2)$ held. The fact that the system is actually 2d (or 3d, see below) does not only lower the dispersion along c by some constant for constant q_b but adds another q_c dependence. The symmetry about $q_c = \pi$, which one has in $d = 1$ due to dimerization [21], is broken by the term proportional to $\cos(q_c)$. Since t_{11} is $\approx 1/7$ of t_{10} the effect is not very large, but visible. Physically this mixture of motion in the b- and c-direction is understandable in fig. 1.

Any hop along b implies a shift also along c because the dimers alternate. In addition, the motion along b lowers the observable gap. Hence a given gap corresponds to a larger dimerization than in $d = 1$.

For a more quantitative comparison of theory and experiment we extend (1) to third order by standard degenerate perturbation theory

$$t_0 = 1 - (4 - 3\bar{\alpha}^2)\lambda^2/16 - (4 - 3\bar{\beta}^2)\mu^2/8 - (8 - 8\bar{\alpha} - 6\bar{\alpha}^2 + 3\bar{\alpha}^3)\lambda^3/64 - (8 - 8\bar{\beta} - 6\bar{\beta}^2 + 3\bar{\beta}^3)\mu^3/32 + (8\bar{\alpha} - 16\bar{\beta} + 18\bar{\alpha}\bar{\beta} + 9\bar{\beta}^2 - 12\bar{\alpha}\bar{\beta}^2)\lambda\mu^2/32 + \lambda^4/128 - 3\lambda^2\mu^2/16 - 3\mu^4/64 \quad (2a)$$

$$t_{10} = -\bar{\alpha}\lambda/4 - \lambda^2/8 - \bar{\beta}^2\mu^2/16 - (4 - 4\bar{\alpha} - \bar{\alpha}^3)\lambda^3/64 - (2 - \bar{\beta})\bar{\beta}^2\mu^3/32 + (6\bar{\alpha} - 6\bar{\beta} + \bar{\beta}^2 + 2\bar{\alpha}\bar{\beta}^2)\lambda\mu^2/32 + \lambda^4/128 + \lambda^2\mu^2/32 + 3\mu^4/128 \quad (2b)$$

$$t_{11} = -\bar{\beta}\mu/4 - \bar{\alpha}\bar{\beta}\lambda\mu/16 - \mu^2/8 + (4\bar{\alpha} + 12\bar{\beta} - 4\bar{\alpha}\bar{\beta} + 7\bar{\alpha}^2\bar{\beta})\lambda^2\mu/128 + (\bar{\alpha} - \bar{\beta})\lambda\mu^2/8 - (8 - 16\bar{\beta} - 7\bar{\beta}^3)\mu^3/128 + 5\lambda^2\mu^2/128 + 3\mu^4/128 \quad (2c)$$

$$t_{20} = -\bar{\alpha}^2\lambda^2/32 - (2 - \bar{\alpha})\bar{\alpha}^2\lambda^3/64 - 3\bar{\alpha}\bar{\beta}^2\lambda\mu^2/64 + 3\lambda^4/256 \quad (2d)$$

$$t_{21} = -\bar{\alpha}\bar{\beta}\lambda\mu/16 - (8\bar{\alpha} - 8\bar{\beta} + 8\bar{\alpha}\bar{\beta} + 5\bar{\alpha}^2\bar{\beta})\lambda^2\mu/256 + (2\bar{\alpha} - 2\bar{\beta} - 2\bar{\alpha}\bar{\beta} + \bar{\alpha}\bar{\beta}^2)\lambda\mu^2/64 - 3\bar{\beta}^3\mu^3/256 + 3\lambda^2\mu^2/128 \quad (2e)$$

$$t_{22} = -\bar{\beta}^2\mu^2/32 - 3\bar{\alpha}\bar{\beta}^2\lambda\mu^2/64 - (2 - \bar{\beta})\bar{\beta}^2\mu^3/64 + 3\mu^4/256 \quad (2f)$$

$$t_{23} = -\bar{\beta}^2\mu^2/16 - 3\bar{\alpha}\bar{\beta}^2\lambda\mu^2/64 + (4 - 2\bar{\beta} + \bar{\beta}^2)\bar{\beta}\mu^3/32 + 3\mu^4/128 \quad (2g)$$

$$t_{30} = -\bar{\alpha}^3\lambda^3/128 \quad t_{31} = -3\bar{\alpha}^2\bar{\beta}\lambda^2\mu/128 \quad (2h)$$

$$t_{32} = -3\bar{\alpha}\bar{\beta}^2\lambda\mu^2/128 \quad t_{33} = -\bar{\beta}^3\mu^3/128 \quad (2i)$$

$$t_{34} = -3\bar{\beta}^3\mu^3/128. \quad (2j)$$

All frustration terms have been included with $\bar{\alpha} = 1 - 2\alpha$ and $\bar{\beta} = 1 - 2\beta$. This is not too difficult once one has the perturbation terms for $\alpha = 0, \beta = 0$ because the frustration terms link the same dimers and they have the same effects on them except for the matrix elements. The dispersion reads

$$\begin{aligned} \frac{\omega(\vec{q})}{J} = & [t_0 + 2t_{10} \cos(2q_c) + 2t_{20} \cos(4q_c) + 2t_{30} \cos(6q_c) \\ & + 4 \cos(q_b/2)(t_{11} \cos(q_c) + t_{21} \cos(3q_c) + t_{31} \cos(5q_c)) \\ & + 2 \cos(q_b)(t_{23} + 2t_{22} \cos(2q_c) + 2t_{32} \cos(4q_c)) \\ & + 4 \cos(3q_b/2)(t_{34} \cos(q_c) + t_{33} \cos(3q_c))]. \end{aligned} \quad (3)$$

For $\mu = 0, \alpha = 0$, (2), (3) can be checked with the result for a dimerized NN chain [22]. For $\mu = 0, \lambda\alpha = J/J_\perp, \lambda = 0$ one retrieves the result for a NN spin ladder [23]. The upper chain is made up by the first sites of the dimers; the lower chain of the second sites. For $\mu = 0, \lambda\alpha = J/J_\perp$ and arbitrary λ , a general zigzag chain is described. This should give an excellent description of the magnon dispersion in $\text{Cu}_2(\text{C}_5\text{H}_{12}\text{N}_2)_2\text{Cl}_4$ [24].

Another special case is $((\alpha = 1/2 \text{ or } \lambda = 0) \text{ and } (\beta = 1/2 \text{ or } \mu = 0))$ where brackets, ‘ands’, and ‘ors’ are logical signs. The singlet product state is the ground state in the $d = 1$ case $\mu = 0, \lambda \leq 1$ [25]. The same can be proven for the $d = 2$ lattice in fig. 2 if $\alpha = \beta = 1/2$. For this case the fourth order terms are included in (2). It is reasonable to include the fourth order terms if α, β are close to $1/2$.

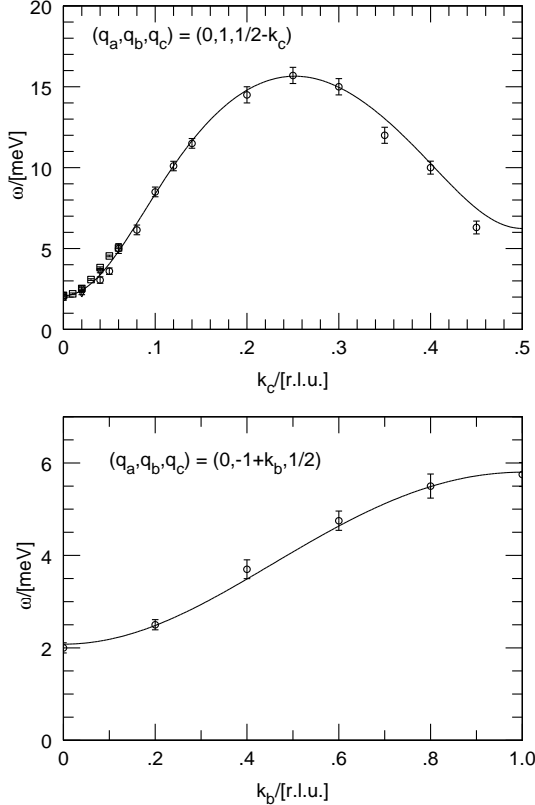


FIG. 3. a: Dispersion along the c-axis resulting from (2) and (3) at $(J, \lambda, \alpha, \mu, \beta) = (11.0\text{meV}, 0.78, 0, 0.30, 0.3)$ with experimental data after [9], fig. 8. Wave vectors in reciprocal lattice units (1 r.l.u. = $2\pi/\text{lattice spacing}$). b: Same as in fig. 3a along the b-axis (after [9], fig. 5).

In fig. 3, a comparison between the theory and INS results is made. The best fit is obtained for $\alpha = 0$ although recent excellent $\chi(T)$ -fits indicate that $\alpha = 0.35$ for $T > 30$ K is realistic [26,27]. The discrepancy can be due to the truncation of the perturbation series at third order or to a change of α across the spin-Peierls transition induced by the static distortion or induced by a renormalization because of non-adiabatic phonon effects. This remains to be clarified.

For the $\alpha = 0$ fit the terms $\propto \lambda^2 \mu^2, \lambda^4$ in (2) are dropped. The value $\lambda = 0.78$ corresponding to $\delta = 0.12$ is not as small as assumed previously ($\delta = 0.03 \dots 0.05$ [11–13,9]) but is of the order of ‘first principles’ values $\delta = 0.21$ [20] or $\delta \approx 0.1$ [28]. Note that the inclusion of higher order perturbation terms, enhancing $\omega_{\text{max}} - \omega_{\text{min}}$ at constant λ , will lower the appropriate fit value for λ even further.

The dispersion along the a-axis is accounted for by adding to $\omega(\vec{q})$ as given by (3) a term $\omega_a(\vec{q}) = 4t_a \cos(q_a) \cos(q_c)$ with $4t_a = -0.22\text{meV}$ [6]. This term is analogous to the third term in (1) since the dimerization pattern alternates also in a-direction [19].

The agreement is excellent in fig. 3a for $q_c < 0.3$, where most data points are available, and it is still very good for higher values. The data points show clearly a small shift to higher values of q_c which results from the terms proportional to $\cos(nq_c)$ with n odd. Such terms are a signature of interchain hopping with alternating dimerization. The agreement in fig. 3b is also good.

The fit in fig. 3 corroborates the approach to describe the dispersion as hopping on the lattice in fig. 2. The parameters, however, $(J, \lambda, \alpha, \mu, \beta)$ are not unambiguously fixed by the dispersion data. Good fits can also be obtained for smaller values of μ and β , e.g. $(J, \lambda, \alpha, \mu, \beta) = (10.7\text{meV}, 0.78, 0, 0.14, 0)$. This can be understood by the fact that the main hopping effect along b is $\propto \mu(1 - 2\beta)$. To fix the value of β we deduced the coefficients $\chi_1 \approx -7.8 \pm 0.9\text{meV}$ and $\chi_2 \approx 26 \pm 7\text{meV}^2$ in a large T-expansion of the magnetic susceptibility $\chi(T) = (\chi_0/T)(1 + \chi_1/T + \chi_2/T^2 + \dots)$ from the data in [1]. Performing this expansion for the undimerized counterpart of the lattice in fig. 1 yields

$$\chi_1 = -(J_\lambda + J_\mu + J_\alpha + 2J_\beta)/2 \quad (4a)$$

$$\chi_2 = (J_\beta^2 + J_\lambda(J_\mu + J_\alpha) + 2J_\beta(J_\alpha + J_\mu + J_\lambda))/2 \quad (4b)$$

where $J_\lambda = (1 + \lambda)J/2$, $J_\alpha = J\lambda\alpha$, $J_\mu = J\mu$, and $J_\beta = J\mu\beta$. For parameters as in fig. 3, one has $\chi_1 = -7.5\text{meV}$, $\chi_2 = 30\text{meV}^2$ while for $\beta = 0$ one has $\chi_1 = -5.4\text{meV}$, $\chi_2 = 6.9\text{meV}^2$. This points to a certain amount of interchain frustration β not too far away from $\beta \approx 0.5$.

Fig. 4 displays the overall form of $\omega(\vec{q})$ differing from the schemes in [18,4]: a) In fig. 4 $\omega(\vec{q})$ is roughly constant for $q_c = 0.25$, i.e. close to the maximum value of $\omega(\vec{q})$, whereas it displays the same dependence on q_b at all q_c in [18,4]. b) According to [18,4] the smaller gap $E_{g1} \approx 2.1\text{meV}$ is found at $\vec{q} = (0, 1, 1/2)$ and $(0, 1, 0)$; the larger gap $E_{g2} \approx 5.5\text{meV}$ is found at $\vec{q} = (0, 0, 1/2)$ and $(0, 0, 0)$. In fig. 4, E_{g1} and E_{g2} are swapped at $q_c = 0$. This is very important in the interpretation of experimental data (INS, Raman, ESR, FIR). The magnon DOS deduced from fig. 4 is constant at the lower band edge (2d minimum), it has a logarithmic singularity at E_{g2} (2d saddle point), but resembles to a 1d, $1/\sqrt{\omega_{\text{max}} - \omega}$ DOS at the maximum due to the approximate degeneracy at $q_c = 0.25$.

Magnetic resonance data [29,30] shows an excitation from the ground state at E_{g2} which was interpreted to be the gap at $(0, 0, 0)$. But a vertical transition at $\vec{q} = (0, 0, 0)$ from $S = 0$ (ground state) to $S = 1$ (excited states) is actually impossible due to total spin conservation.

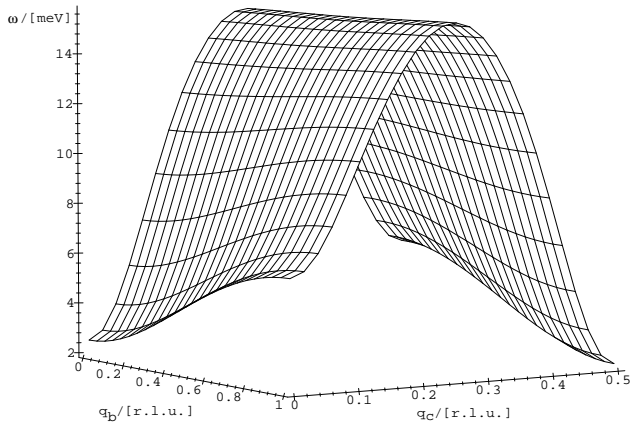


FIG. 4. 3D representation of the dispersion $\omega(\vec{q})$ for $q_a = 0$ (parameters as in fig. 3).

For this reason and in view of fig. 4, a re-interpretation of this resonance is proposed. In analogy to the magnetic resonances due to transitions ground state \rightarrow excited state in NENP and NINO (see e.g. [31,32]) the transition reflects the situation at $(0, 1, 0)$ [33] because of the presence of staggered magnetic fields due to differently orientated g -tensors on neighboring chains in b -direction [34]. The Cu-atoms in each unit cell are inequivalent since their crystal environments are differently orientated [20] (cf. the situation for NENP/NINO [35]). This argument makes the magnetic resonance data compatible with fig. 4 (within 10% accuracy as for NENP [32]).

Further INS investigation are desirable to make a more precise analysis possible. It should be noted that the dispersion along the a -axis leads to *four* different gaps in fig. 4. For the parameter set used in figs. 3 and 4, one has $\omega((0, 1, 1/2)) = 2.1\text{meV}$, $\omega((0, 0, 0)) = 2.5\text{meV}$, $\omega((0, 0, 1/2)) = 5.8\text{meV}$, and $\omega((0, 1, 0)) = 6.2\text{meV}$.

The magnon dispersion described will, at least qualitatively, also apply to the currently investigated α' - NaV_2O_5 where the dimerization is equally alternating from chain to chain [3]. Such an alternation appears to be generic for inorganic substances where bond angles are more easily changed than bond lengths.

In summary, we showed that the lattice distortion in the spin-Peierls phase causes a dispersion which corresponds to hopping on a lattice as in fig. 2. Thus the gap positions in the Brillouin zone are revisited. A general perturbative result is given to enable a quantitative analysis of the dispersion. A larger dimerization ($\delta \approx 0.1$) in agreement with orbital calculations is found. This result covers also chains and certain ladders which are a very active field as well [36]. The magnetic resonance results are reinterpreted.

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